

# First direct measurements on $^{56}\text{Ni}$ and $^{59}\text{Ni}$ with fast neutrons at LANSCE

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03/01/2022

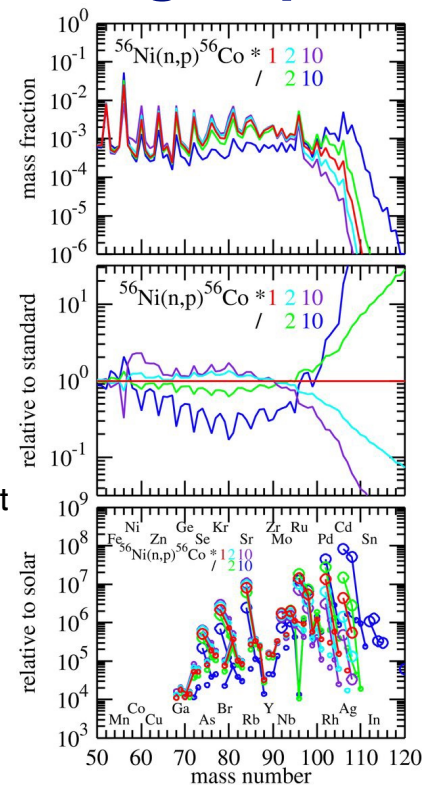
# Outline

- Neutron induced charged particle measurements with LENZ at LANSCE
- Study of  $^{59}\text{Ni}(n,p)^{59}\text{Co}$  and  $^{59}\text{Ni}(n,\alpha)^{56}\text{Fe}$  with a  $^{59}\text{Ni}$  target
  - Comparison to a surrogate ratio measurement
  - $^{59}\text{Ni}$  is a significant background component to our measurement of  $^{56}\text{Ni}(n,p)$
- Study of  $^{56}\text{Ni}(n,p)^{56}\text{Co}$  and  $^{56}\text{Co}(n,p)^{56}\text{Fe}$  with a radioactive Ni/Co cocktail target
- Development of a solenoid spectrometer for improved measurements with radioactive targets.
- Summary/outlook

# Nuclear data needs for neutron-induced charged-particle reactions (n,z)

- Damage due to hydrogen and helium production in structural materials like Fe, Cr, Ni, etc.
  - Manuscript on  $^{54}\text{Fe}(n,z)/^{56}\text{Fe}(n,z)$  to be submitted for publication.
  - Measurements of  $^{58}\text{Ni}(n,z)/^{60}\text{Ni}(n,z)$  with LENZ are under analysis (D. Votaw)
- Precision measurements of key reactions like  $^6\text{Li}(n,t)^4\text{He}$ ,  $^{10}\text{B}(n,\alpha)^7\text{Li}$ ,  $^{12}\text{C}(n,\alpha)^9\text{Be}$ ,  $^{16}\text{O}(n,\alpha)^{13}\text{C}$ , etc.
  - Kuvin et al. Phys. Rev. C, 104, 014603 (2021)
  - Manuscript on  $^{16}\text{O}(n,\alpha)^{13}\text{C}$  to be submitted for publication.
- Informing the design of next-gen reactions (e.g. fast spectrum molten salt reactors) where reactions like  $^{35}\text{Cl}(n,p)^{35}\text{S}$  can play a significant role as a neutron poison and produces  $^{35}\text{S}(T_{1/2} \sim 75 \text{ days})$  that can complicate the path to certification for designs that incorporate chloride salts.
  - Study of  $^{35}\text{Cl}(n,p)^{35}\text{S}$ : S Kuvin et al. Phys. Rev. C, 102, 024623 (2020)
- Constraining the vp-process for nuclear astrophysics by studying (n,p) reactions on proton-rich unstable nuclei (radioactive targets). e.g.  $^{56}\text{Ni}(n,p)^{56}\text{Co}$  ( $^{56}\text{Ni}$   $T_{1/2} \sim 6 \text{ days}$ )
- Radiochemistry diagnostics for quantifying performance of nuclear fuel burning.

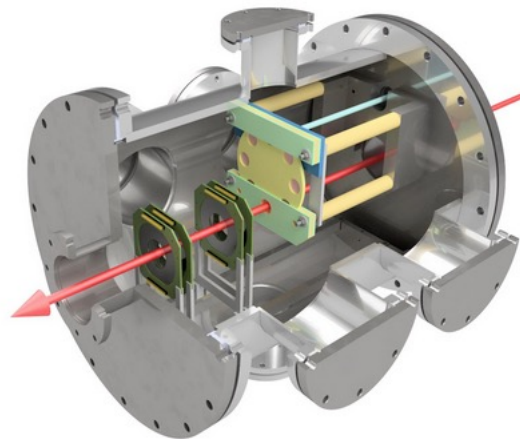
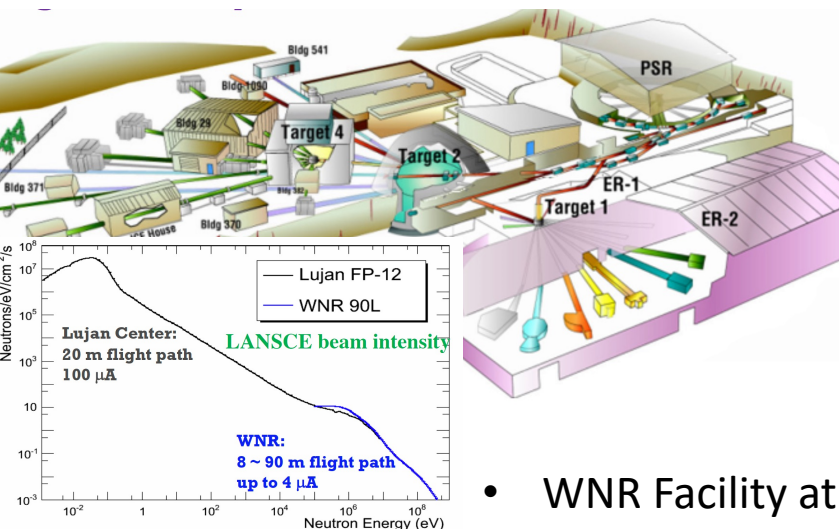
LENZ: Low Energy (n,z) collaboration/experimental setup developed to pin down these types of reactions that are ubiquitous in nature.



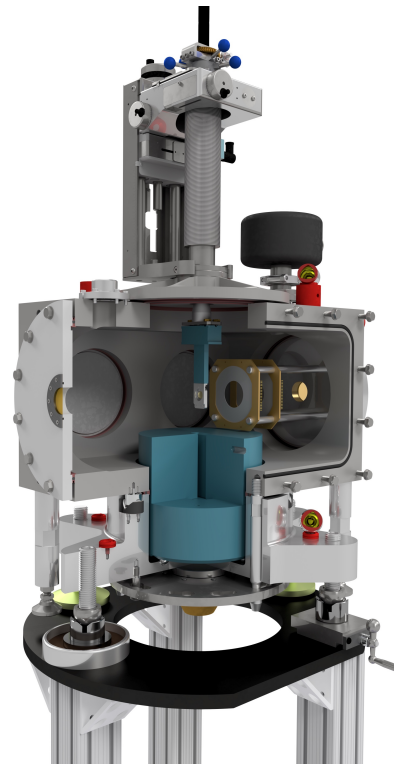
As the most abundant seed nucleus for the vp-process, any small change in the  $^{56}\text{Ni}(n,p)^{56}\text{Co}$  rate results in a significant impact on final abundances of heavier isotopes (Wanajo et al)

# LENZ: The Low Energy (n,z) experimental station

- Detect outgoing charged particles using double-sided silicon strip detectors in a compact setup close to the target sample.



*Schematic diagram of the LENZ instrument, composed of two sets of dE DSSD detector telescopes at forward angles, and a target wheel in the middle of the instrument. Red arrow shows the neutron beam direction.*



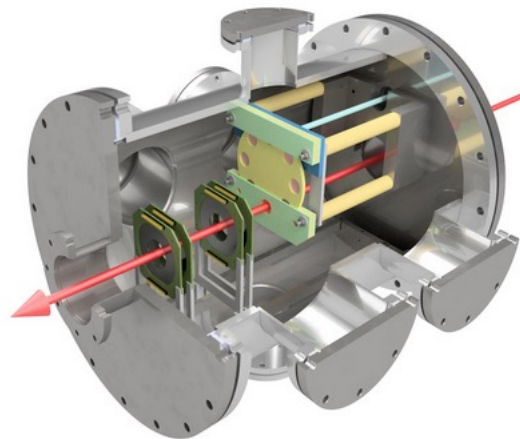
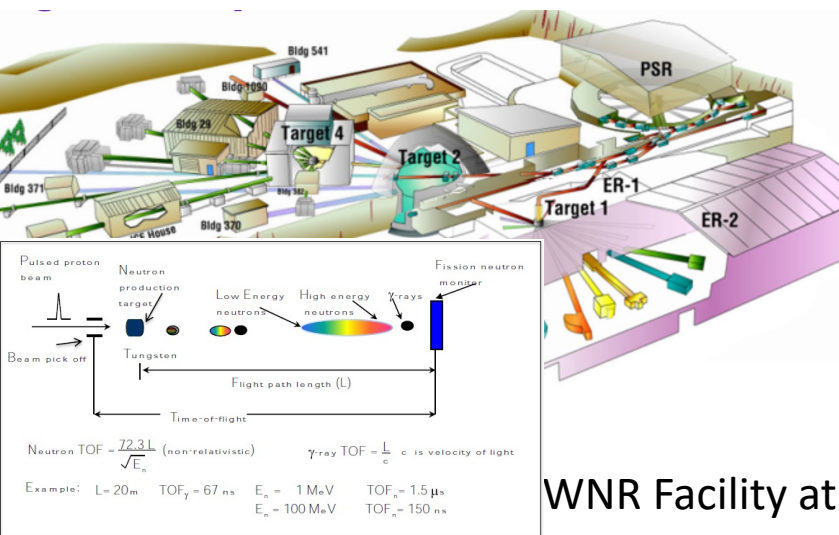
hotLENZ  
rendering by B.  
DiGiovine  
3/1/22

- WNR Facility at LANSCE: fast neutrons with a broad energy spectrum  $\sim 100$ s of keV to  $\sim 100$ s of MeV

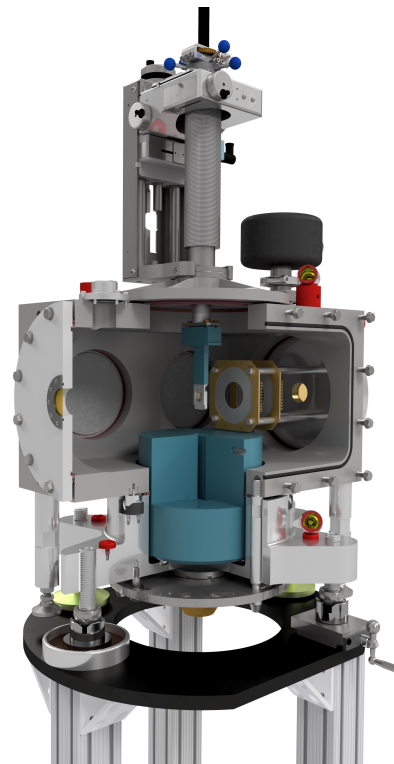


# LENZ: The Low Energy (n,z) experimental station

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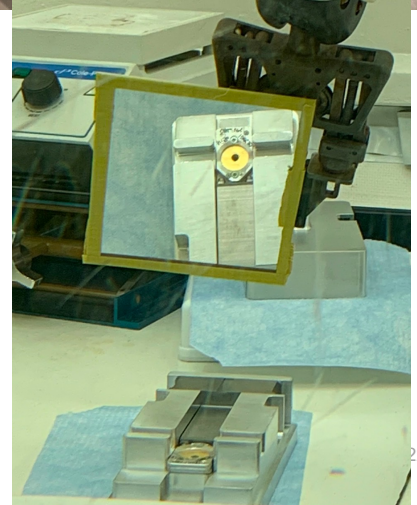
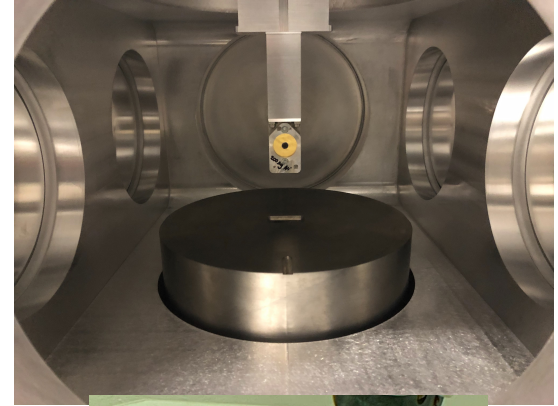


hotLENZ  
rendering by B.  
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WNR Facility at LANSCE: fast neutrons with a broad energy spectrum  $\sim 100\text{s of keV}$  to  $\sim 100\text{s of MeV}$

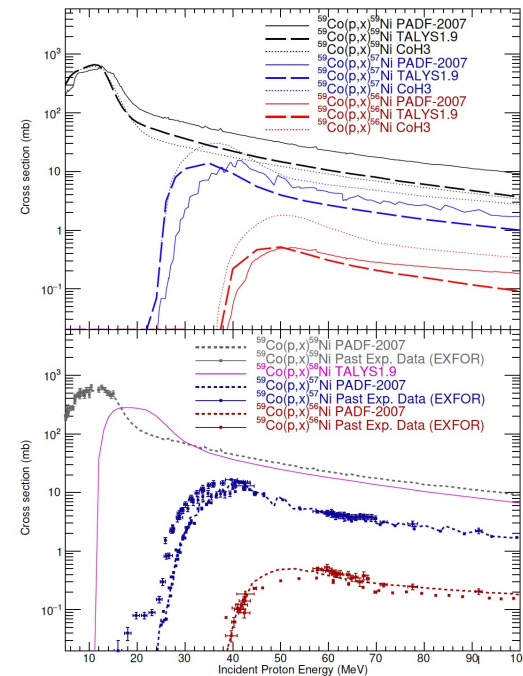
# hotLENZ at WNR and target fabrication at IPF

- Goal: Leverage a world unique capability that exists at LANSCE for target production at the Isotope Production Facility, purification and fabricated at the Isotope Program Hot Cell Facility, and then studied using the fast neutron beams available at the WNR facility.
- See talk by B. DiGiovine for more details on the infrastructure improvements and engineering behind hotLENZ and the talk by V. Mocko for more details on the IPF Hot Cell Facility and the chemical separation and target fabrication efforts.
- The efforts that they will describe made this work, and other recent radioactive target experiments at LANSCE, possible.

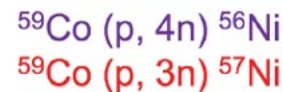


# Production of $^{56}\text{Ni}$ at IPF via $^{59}\text{Co}(p,x)$

- The production of  $^{56}\text{Ni}$  via  $^{59}\text{Co}(p,xn)$  reactions, also results in the production of  $^{57}\text{Ni}$ ,  $^{58}\text{Ni}$ , and  $^{59}\text{Ni}$  that will all be present in the final radioactive cocktail target, without performing further mass separation. Since  $^{56}\text{Ni}$  and  $^{57}\text{Ni}$  will decay swiftly with day-long half-lives, whereas  $^{58}\text{Ni}$  (stable) and  $^{59}\text{Ni}$  (long lived) will not, the charged particle backgrounds due to  $^{58}\text{Ni}(n,z)$  and  $^{59}\text{Ni}(n,z)$  will be dominant.
- Measurements of the stable nickel isotopes are captured in many past measurements. However, the lack of past experimental data on  $^{59}\text{Ni}(n,z)$  at fast neutron energies above 100 keV, presents a technical consideration for the study of  $^{56}\text{Ni}(n,p)$  that needs to be characterized.

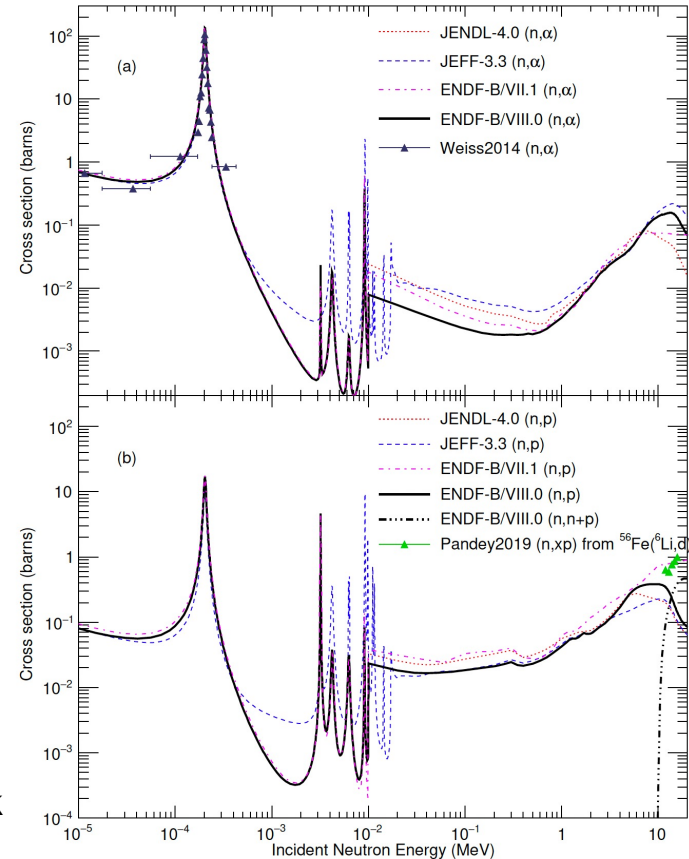


Ni56 5.9 d	Ni57 35.6 h	Ni58 68.1	Ni59 7.6e4 a	Ni60 26.2	Ni61 1.14
$\epsilon$	$\epsilon, \beta^+$		$\epsilon, \beta^+$		
Co55 17.53 h $\beta^+$	Co56 77.3 d $\epsilon, \beta^+$	Co57 271.8 d $\epsilon$	Co58 9.1 h 70.9 d IT $\epsilon, \beta^+$	Co59 100	Co60 10.5 m 5.3 a $\beta^+ \gamma$



# $^{59}\text{Ni}(n,p)^{59}\text{Co}$ and $^{59}\text{Ni}(n, \alpha)^{56}\text{Fe}$

- $^{59}\text{Ni}$  is a long-lived ( $T_{1/2} \sim 100,000$  years) unstable isotope of nickel that is bookended between the stable  $A=58$  and  $A=60$  isotopes.
- Can build up to a non-negligible portion of the total nickel content in reactors from neutron capture on  $^{58}\text{Ni}$  at thermal energies and from  $^{60}\text{Ni}(n,2n)$  at fast neutron energies in fusion reactors.
- No prior experimental data on  $^{59}\text{Ni}(n,p)$  or  $^{59}\text{Ni}(n,\alpha)$  at fast neutron energies above 20 keV, except for  $(n,xp)$  cross sections derived through a surrogate ratio method (SRM) above 10 MeV.
- Measured with LENZ during the 2019 ( $\sim 1$  ug of  $^{59}\text{Ni}$ ) and 2020 ( $\sim 100$  ug  $^{59}\text{Ni}$ , 95% enriched) run-cycles.
- JEFF-3.3 includes the most recent evaluation of  $^{59}\text{Ni}$  from thermal to fast neutron energies.
- New data allows for a sensitive test of calculations that use global input parameters and/or to benchmark indirect methods like SRM.



# New data on $^{59}\text{Ni}(n,p)$ and $^{59}\text{Ni}(n,\alpha)$ at fast neutron energies

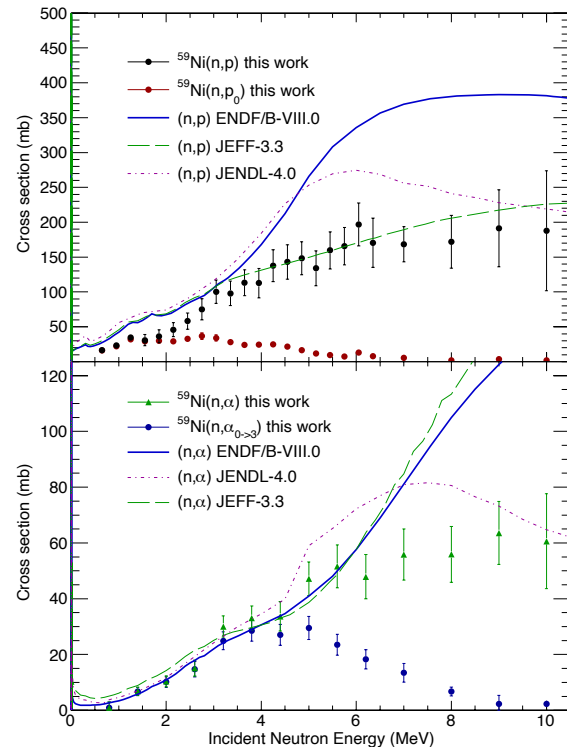
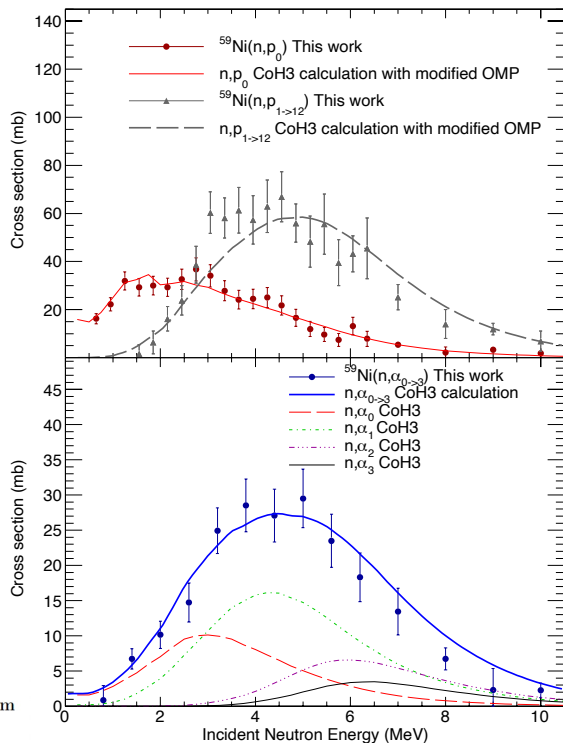
- Statistical Hauser-Feshbach calculations performed using the code CoH3.
- For  $(n,p)$  below 3 MeV, the experimental cross section is approximately 30% lower than the available evaluations and from the calculations when using default parameters. Details on the adjustments to the OMP/level density are described in a publication that is currently under review:

Direct measurement of  $^{59}\text{Ni}(n,p)^{59}\text{Co}$  and  $^{59}\text{Ni}(n,\alpha)^{56}\text{Fe}$  at fast neutron energies from 500 keV to 10 MeV

S. A. Kuvín,<sup>\*</sup> H. Y. Lee, B. DiGiiovine, C. Eiroa-Lledo, A. Georgiadou, M. Herman, T. Kawano, V. Mocko, S. Mosby, C. Vermeulen, D. Votaw, M. White, and L. Zavorka<sup>†</sup>  
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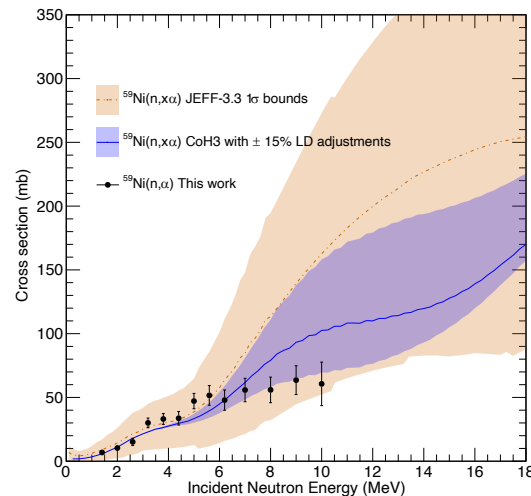
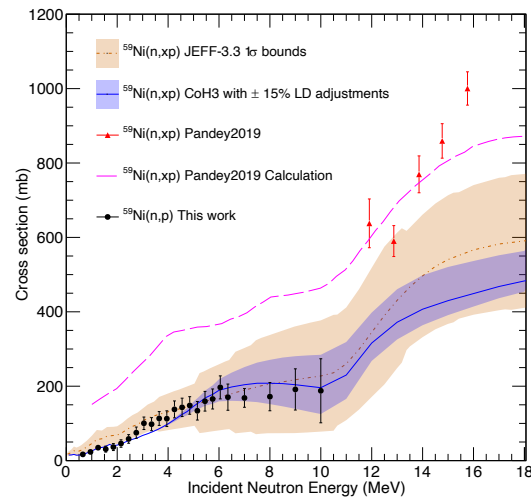
H. I. Kim  
Nuclear Physics Application Research Division, Korean Atomic Energy Research Institute, Yuseong-gu, Daejeon, Korea  
(Dated: February 10, 2022)





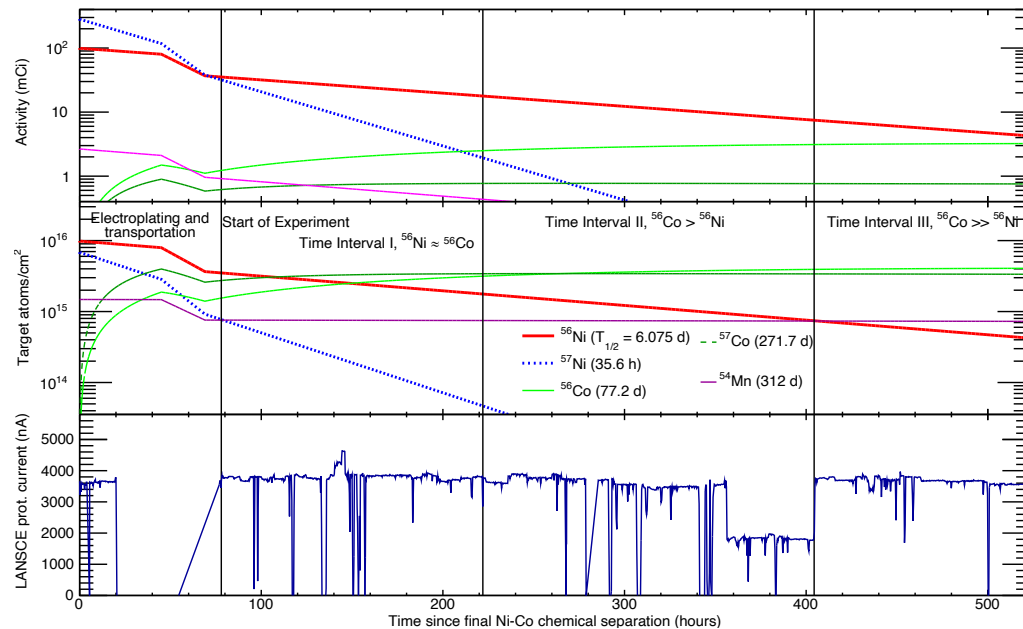
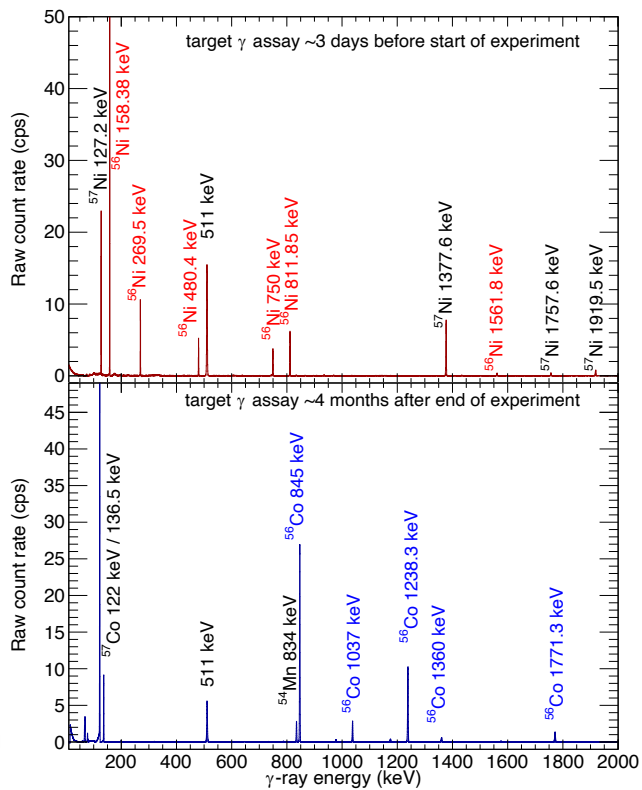
# Study of $^{59}\text{Ni}(n,p)$ and $^{59}\text{Ni}(n,\alpha)$

- Going from the upper to lower bounds of the  $1\sigma$  band from JEFF-3.3 represents a range of nearly a factor of 5 and is inconsistent with the cross sections derived from the surrogate work.
- Our direct measurement is in fairly good agreement with the central value of JEFF-3.3 above  $E_n = 3$  MeV but requires a slight adjustment below 3 MeV.
- Raises questions about the reliability of that particular application of the SRM.
- Direct measurements on radioactive isotopes should be made, when feasible.



# First direct measurement of neutron induced reactions on $^{56}\text{Ni}$ (and $^{56}\text{Co}$ )

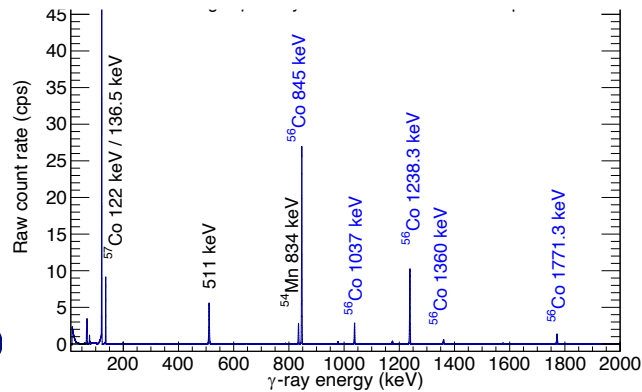
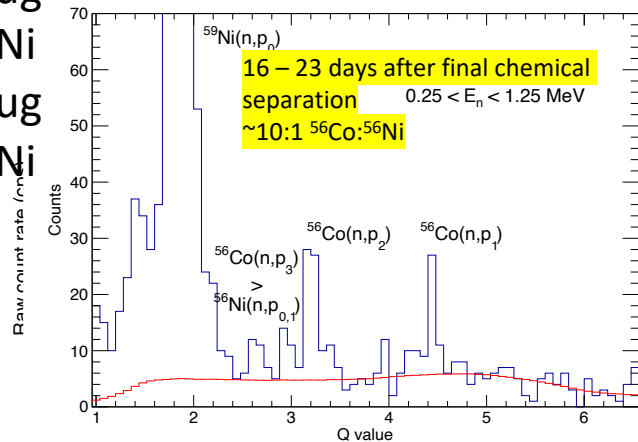
- Approach: Work backwards by analyzing the  $^{56}\text{Co}(n,p)$  at the end of the experiment ( $\sim 10:1$   $^{56}\text{Co}:^{56}\text{Ni}$ ) to determine the  $^{56}\text{Co}$  contributions at the beginning of the experiment when  $^{56}\text{Co}:^{56}\text{Ni}$  was  $\sim 1:1$ .



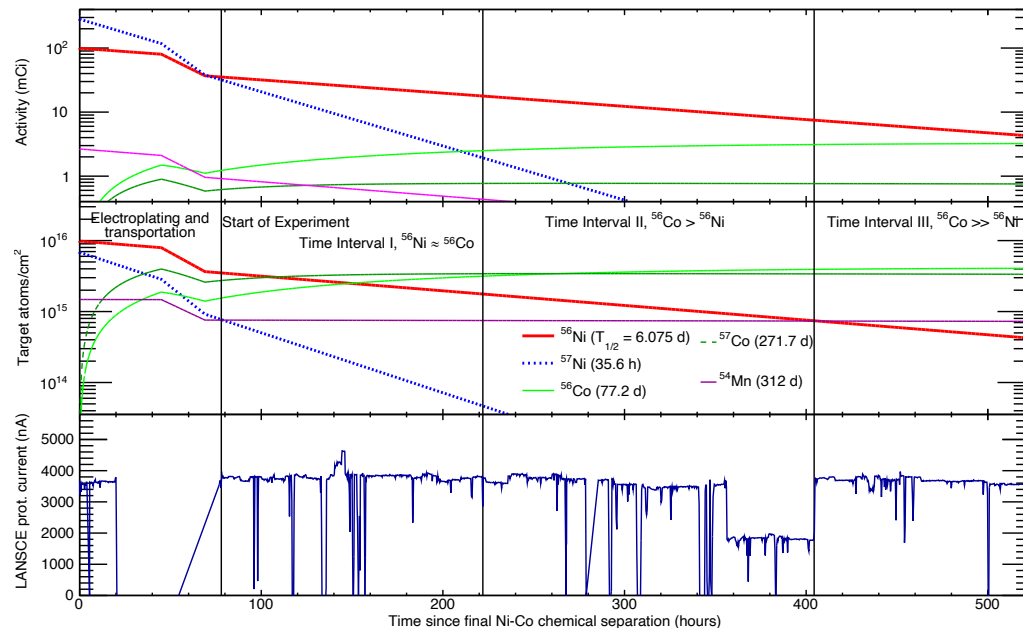


# First direct measurement of neutron induced reactions on $^{56}\text{Ni}$ (and $^{56}\text{Co}$ )

~25 ug  
of  $^{59}\text{Ni}$   
~75 ug  
of  $^{58}\text{Ni}$

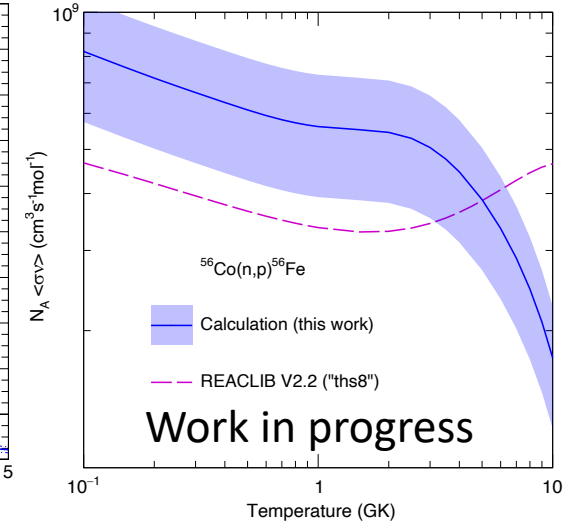
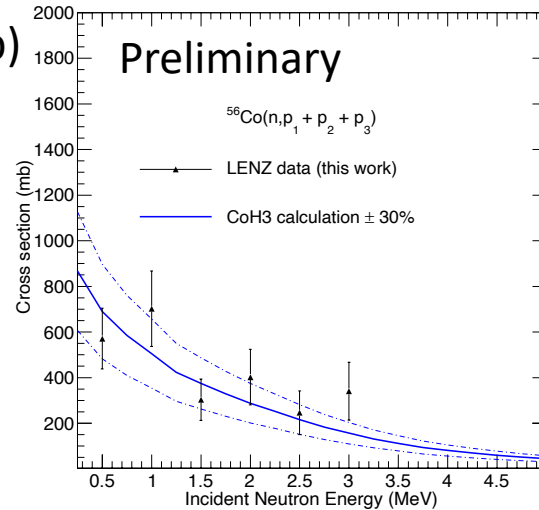


- Approach: Work backwards by analyzing the  $^{56}\text{Co}(n,p)$  at the end of the experiment (~10:1  $^{56}\text{Co}:^{56}\text{Ni}$ ) to determine the  $^{56}\text{Co}$  contributions at the beginning of the experiment when  $^{56}\text{Co}:^{56}\text{Ni}$  was ~1:1.



# First direct measurement of neutron induced reactions on $^{56}\text{Ni}$ (and $^{56}\text{Co}$ )

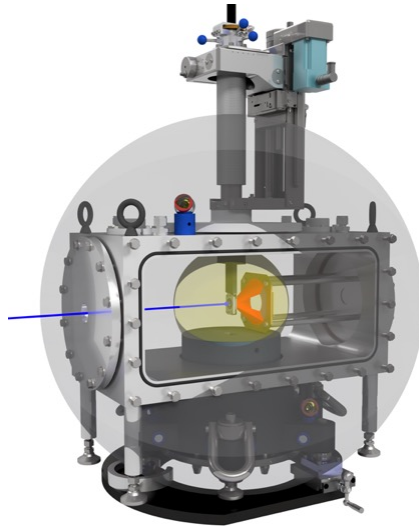
- First experimental data on  $^{56}\text{Co}(n,p)$  in comparison with statistical calculations.
- Preliminary cross sections for  $^{56}\text{Ni}(n,p)$  have been obtained and will provide the first experimental constraints on the role of  $^{56}\text{Ni}(n,p)$  in the vp-process. Stay tuned!
- Total uncertainties are  $\sim 30\%$
- In the process of finalizing the results and the implications of the data for nuclear astrophysics where reaction rates are typically varied by x10-x100 when no experimental data exists



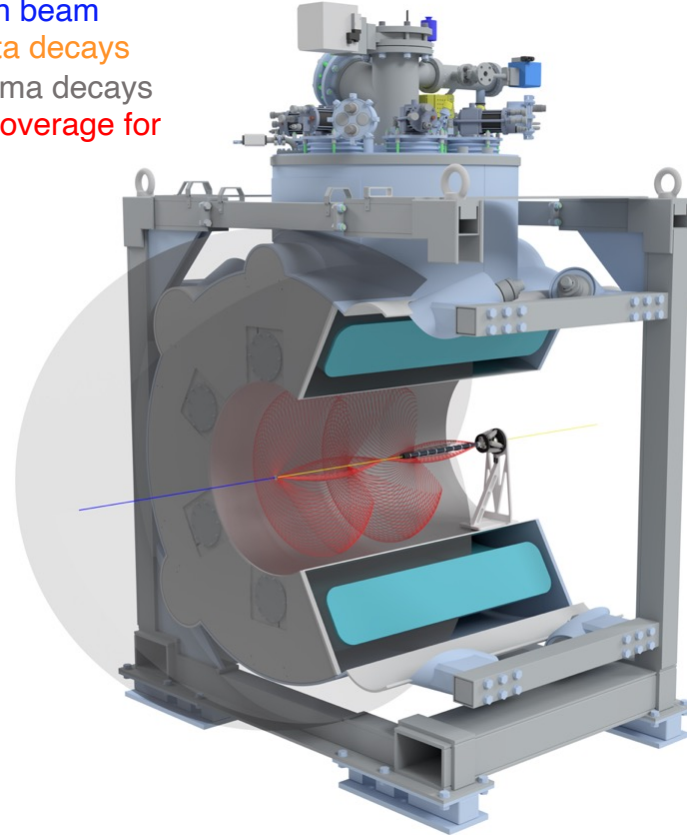
# Solenoid approach for improved measurements with radioactive targets

Blue line: LANSCE neutron beam  
Yellow sphere: intrinsic beta decays  
Gray sphere: intrinsic gamma decays  
Red envelop: solid angle coverage for reaction charged particles

Conventional measurement



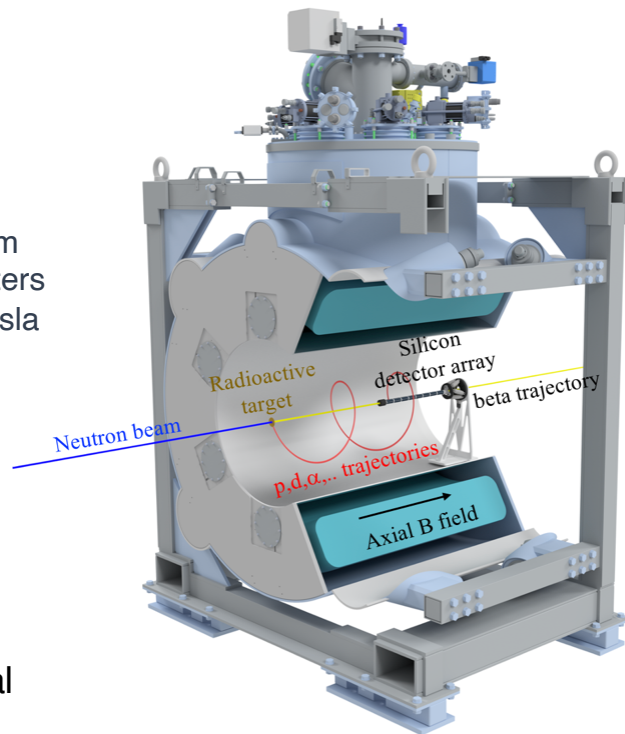
Solenoid approach



# 5T magnet available at LANSCE



Bore diameter = 90 cm  
Bore length = 1.4 meters  
Magnetic Field = 5 Tesla



- Reduced radiation damage to detectors for improved experimental resolutions
- Improved solid angle coverage for charged particles of interest
- Different charged particles are identified by cyclotron period
- Provide high quality nuclear data for reactions with radioactive samples

# Summary/Outlook

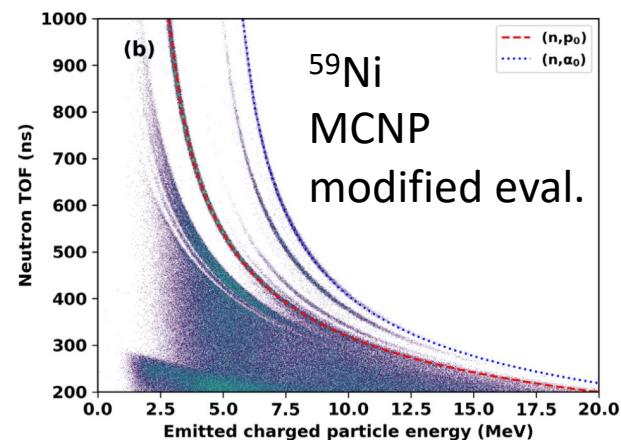
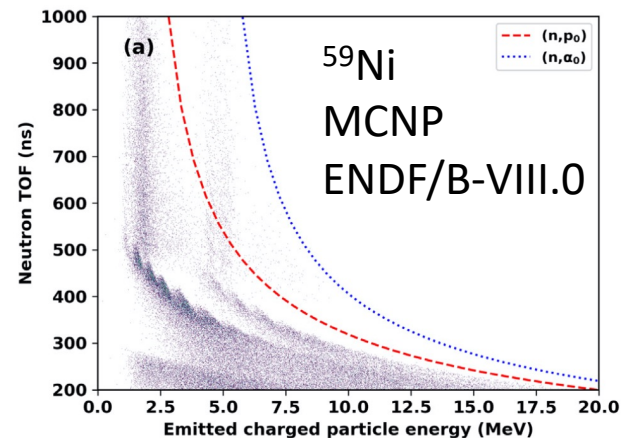
- Study of  $^{59}\text{Ni}(n,p)$  via a direct measurement with a radioactive nickel target demonstrated the importance of making direct measurements, when feasible (manuscript under review with PRC).
- Preliminary cross sections for reactions on radioactive  $^{56}\text{Ni}$  ( $T_{1/2} \sim 6$  days) and  $^{56}\text{Co}$  ( $T_{1/2} \sim 77$  days) have been obtained and we are currently working to finalize the details of the analysis.
- Additional measurements on the stable isotopes  $^{58}\text{Ni}$  and  $^{60}\text{Ni}$  (analysis led by D. Votaw) with LENZ will help provide a more complete evaluation of the nickel isotopes when combined with the radioisotope data.
- In anticipation of some of the nuclear data needs for  $(n,z)$  reactions on unstable nuclei, we have begun planning the development of a solenoidal spectrometer for  $(n,z)$  studies at LANSCE that will have significantly improved sensitivity over a more “traditional approach” to charged particle detection.

# Backup

# Improved evaluations of (n,p) and (n,a)

- Include missing particle production spectrum, discrete states, angular information. ENDF/B-VIII.0 total cross-sections unmodified (and partial cross-sections when available)
- Modified evaluations for (n,p) and (n,a) were performed for 62 different isotopes by Hyeong-il Kim of KAERI and incorporated into the MCNP simulation.

H. I. Kim et al., Nucl. Instrum. Methods Phys. Res. A 964,163699 (2020).  
<https://doi.org/10.1016/j.nima.2020.163699>



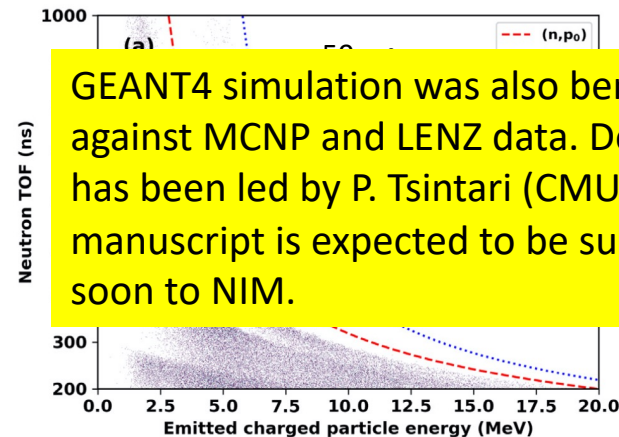
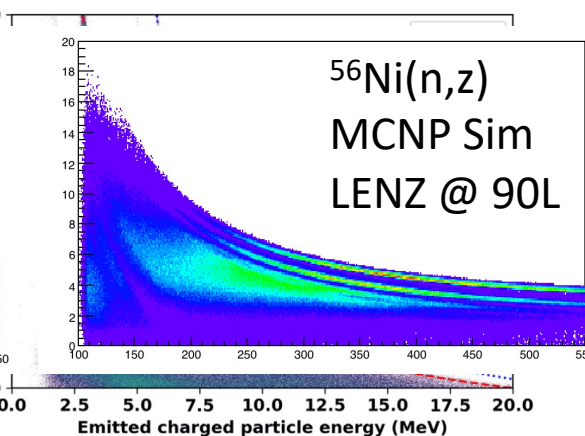
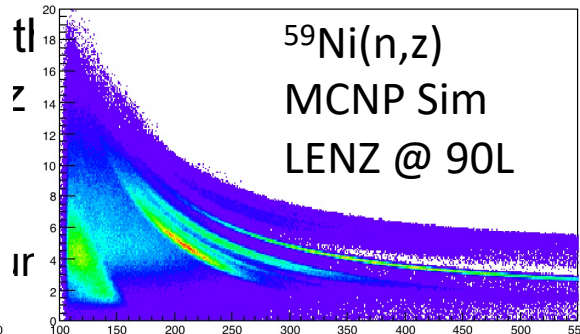
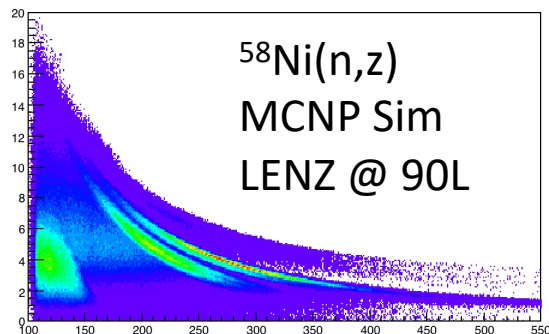


# Improved evaluations of (n,p) and (n,a)

- Include missing particle production spectrum, discrete states, angular information ENDF/B-VIII.0 total

Data libraries also generated for reactions on  $^{56}\text{Ni}$ ,  $^{57}\text{Ni}$ ,  $^{56}\text{Co}$ ,  $^{57}\text{Co}$ , etc for which no previous ENDF/B-VIII.0 evaluation existed

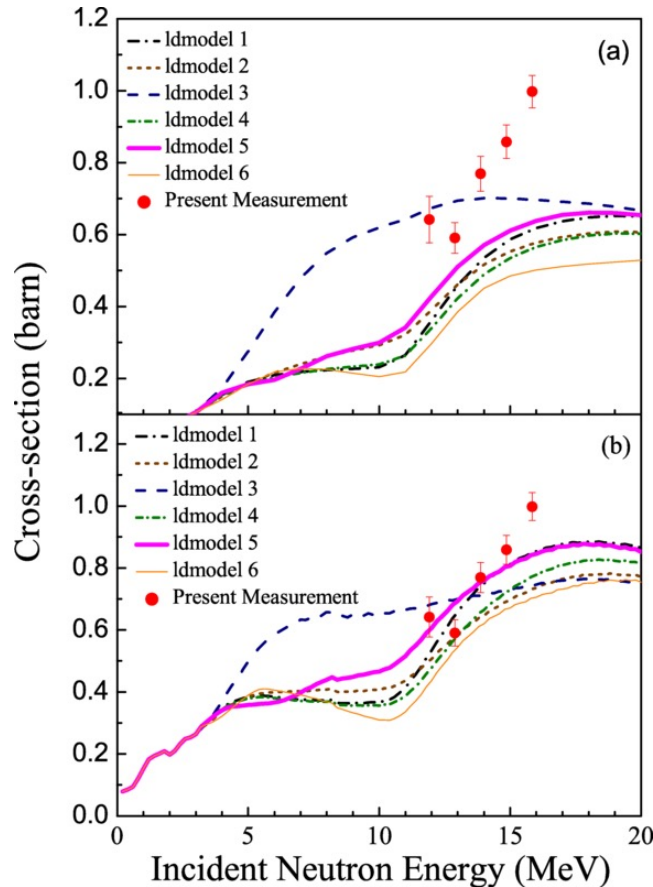
- Modified evaluations for (n,p) and (n,a) were performed for 62 different isotopes by Hyeon-gil Kim of KAERI



GEANT4 simulation was also benchmarked against MCNP and LENZ data. Development has been led by P. Tsintari (CMU). A manuscript is expected to be submitted soon to NIM.

<https://doi.org/10.1016/j.nima.2020.163699>

# Surrogate ratio measurement of $^{59}\text{Ni}(n,xp)$

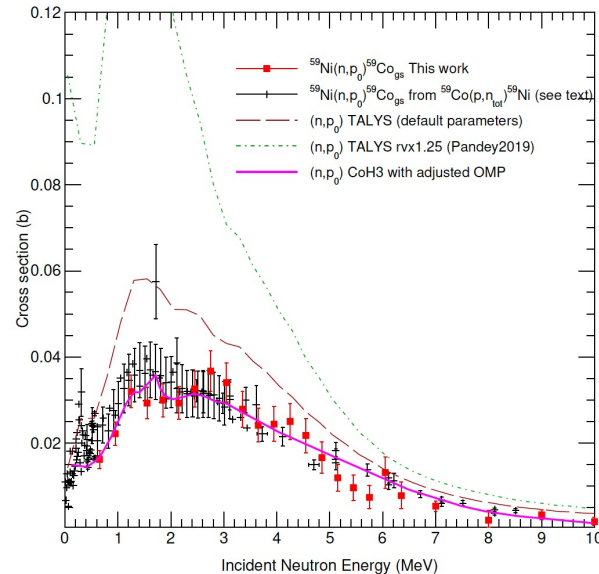
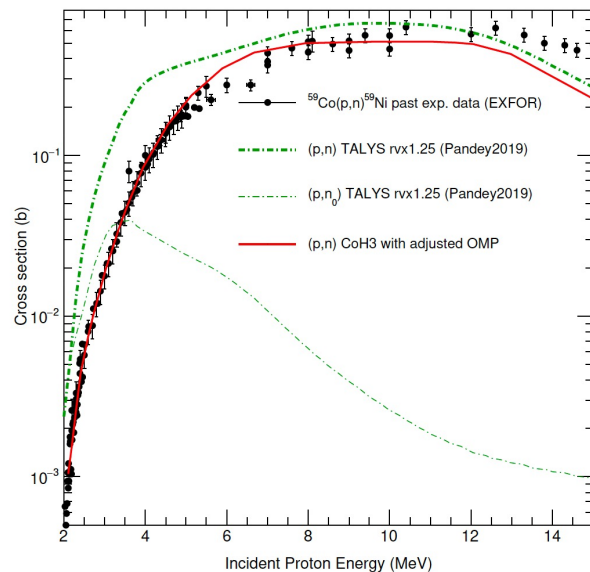


- Prior to this work, the only available  $^{59}\text{Ni}(n,xp)$  cross section information at fast neutron energies was derived from a surrogate ratio method.
- The scale and trend of their data was inconsistent with statistical calculations using default parameters and with ENDF, JEFF, and JENDL for which they conclude that new evaluations are necessary.
- Modifications to the optical potentials used in the statistical calculations are proposed to reproduce the scale of their experimental data.

# Comparison between $^{59}\text{Ni}(n,p)^{59}\text{Co}$ and $^{59}\text{Co}(p,n)^{59}\text{Ni}$

The prescribed adjustment (scaling the volume radius term by 1.25) to the default optical model parameters in TALYS by Pandey *et al.* also performs worse compared to the default parameters when reproducing the  $^{59}\text{Co}(p,n)$  data. It results in a factor 4-5 difference between our direct measurement data and the calculation for  $^{59}\text{Ni}(n,p_0)$  at  $\sim 2$  MeV.

In contrast, we obtain better agreement between the  $(p,n)$  data and our experimental  $(n,p)$  data with the more modest adjustments to the proton OMP.



For  $(n,p_0)$  (right), the magenta curve is from the CoH calculation using the same optical model parameters as the red curve for  $^{59}\text{Co}(p,n)$  case (left). The black data points (right) are derived from the  $^{59}\text{Co}(p,n)$  data from exfor by using the statistical model calculation to get the expected ratio of  $(p,n_0)$  to  $(p,n)$  and then using detailed balance theorem to go from  $(p,n_0)$  to  $(n,p_0)$